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Analysis of Data from the Collocated Operation of Four Radiance Research Nephelometers at Angiola After the End of the CRPAQS Field Study

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ABSTRACT

The configuration of the Radiance Research Nephelometer installations used in the California Regional PM_{10}/$PM_{2.5}$ Air Quality Monitoring Study (CRPAQS) was changed in December 2000 and January 2001. The intercomparison reported here was performed to determine and document the effect of this configuration change on the monitoring data.

During the early part of the study, the relative humidity (RH) sensor was in a fitting on the sample air inlet to the scattering chamber. This configuration allowed the sample airflow to be inadvertently cooled in the scattering chamber, with the result that RH values higher than intended could occur. The configuration was modified by reversing the airflow through the scattering chamber so the RH sensor was on the outlet and by adding thermal insulation to the scattering chamber. Then the heater controller applied heat as necessary to keep the RH at the outlet of the scattering chamber at the intended value.

Two nephelometers in the original configuration and two in the new configuration made collocated measurements from March 2 to 29, 2001 at the Angiola site. Scatter diagrams show that light scattering by particles (b_{sp}) readings from the two nephelometers in the new configuration agreed very well with one another, while readings from the two in the old configuration showed less consistent agreement with each other. Comparison between b_{sp} readings from nephelometers in the old and new configuration showed that when the ambient RH was high enough that the sample air was heated, higher b_{sp} readings were obtained in the old configuration. In the data collected during this intercomparison, the bias is consistent enough that it is possible to apply a linear transformation to convert data obtained using one operating mode into data that would be obtained using the other mode.

Regression analyses were performed to quantify the relations between the b_{sp} measurements recorded when the RH was 66% or less, when the sample airflow heaters were off, at intermediate RH values, and when the RH was 70% or more, when the heaters were on. These data were used to determine the relationship between the nephelometer calibrations and the effect of the nephelometer configuration on the data measured when the heaters were on. An equation was derived that can be used to convert b_{sp} values measured by nephelometers in the original configuration to values that would have been obtained from nephelometers in the new configuration. It is expected that the conversion factors depend on the aerosol composition, so the conversion equation may not be applicable during all seasons of the year.

INTRODUCTION

When Radiance Research Nephelometers were being prepared for use on the Angiola tower in shelters different from those used in most of the monitoring network for CRPAQS, it was noticed that collocated nephelometers gave significantly different readings at times of high RH. An examination of these data led to the conclusion that the standard configuration used for the Radiance Research Nephelometers in CRPAQS was not ideal.

The configuration of the sample airflow system used before December 2000 in CRPAQS had the sample airflow pass through a stainless steel bug screen, through the sample air heater, past the RH sensor, through the nephelometer scattering chamber, and through the exhaust fan that produced the sample airflow. The nephelometer was in a shelter equipped with an exhaust fan that nominally exchanged the air in the shelter every 5 sec. The concern with this configuration is that at low ambient temperatures, the body of the nephelometer is efficiently cooled by the airflow through the shelter. This could cause the temperature in the scattering chamber to be lower and the RH of the air in the chamber to be higher than at the location of the RH sensor on the inlet. The heater controller keeps the RH at the sensor from exceeding a set value, but the RH in the scattering chamber could be higher. A higher RH in the scattering chamber would cause the aerosol to absorb more liquid water and cause the measured b_{sp} to increase. The magnitude of this increase in measured b_{sp} would be expected to increase as the ambient temperature became colder.

This uncontrolled source of variability in the b_{sp} data can be minimized by changing the sample airflow so the RH sensor is on the outlet. Then the heater controller supplies heat as needed to keep the RH at this location at the setpoint, and the RH in the scattering chamber, if anything, is lower. The setpoint of the heater controller was near the RH valve at which aerosol particles deliquesce, so decreasing the RH causes less change in b_{sp} than increasing the RH. Furthermore, the scattering chamber of a nephelometer is approximately well mixed (Bergin et al., 1997) and the distance from the chamber to the RH sensor is very short, so an RH sensor on the outlet should give a reasonably good measure of the RH in the scattering chamber.

The simplest way of moving the RH sensor to the outlet was to reverse the direction of airflow through the nephelometer. In December 2000 it was recommended that the airflow direction through all nephelometers be reversed. This was accomplished by swapping the locations of the sample air fan and sample air heater. In addition, a kit of foam pipe insulation and cable ties was provided for each nephelometer so the body of the nephelometer could be thermally insulated. This minimized the heat loss through the walls of the scattering chamber and hence the amount of heating needed to control the RH at the nephelometer outlet.

This change in operating procedure during the middle of CRPAQS raised questions about the effects of the change on the data. Therefore, an experiment was conducted after the end of the CRPAQS monitoring program to compare the data from two nephelometers with the original configuration with data from two nephelometers with the reversed sample airflow and thermal insulation.

EXPERIMENTAL

The intercomparison study was conducted on the roof of the shelter at Angiola from March 2 to 29, 2001. The ambient temperatures were not as cold and the aerosol concentrations were not as high at this time of year as in the late fall and early winter, so the most extreme conditions encountered during CRPAQS were not explored by this experiment.

Nephelometers with serial numbers 0192 and 0194 were operated in the original configuration, and nephelometers with serial numbers 0262 and 0276 were operated with the reversed sample airflow and thermal insulation. Nephelometer 0276 began operating on March 5 because it replaced a nephelometer that was not operating properly.

Zero and span calibrations were performed for each nephelometer the day each was installed and on March 23 and 29. Zero calibrations were also performed on March 20. The calibration data measured at the start of the intercomparison were mostly outliers; they did not agree with either the results of the most recent calibrations performed during the CRPAQS field study or with the final calibration results obtained on March 20, 23, and 29. Therefore, only the final calibration data were used to calculate the zero and span values in **Table 1**.

Table 1. Zero and span values applied to the measured b_{sp} data.

Nephelometer Serial No.	Zero (Mm^{-1})	Span/Expected Span
0192	3.09	1.007
0194	3.97	0.990
0262	5.22	1.045
0276	0.81	1.054

The standard deviations of the values averaged to obtain the zero for each nephelometer ranged from 0.028 to 0.083 Mm^{-1} and averaged 0.062 Mm^{-1}. The zero measured at the start of the intercomparison that was closest to the zero for that nephelometer in Table 1 differed from the zero in Table 1 by 8 to 14 times the standard deviation of the zero values averaged to obtain the values in Table 1.

Only two spans were performed near the end of the intercomparison. The differences in the span/expected span ranged from 0.005 to 0.017 and averaged 0.011. This corresponds to a 1.1% difference between the two spans. For three nephelometers, the span/expected span measured at the start of the intercomparison differed from the value in Table 1 by 6 to 10 times the difference between the two final spans. In one case, the initial span differed by only 1.5 times the difference between the two final spans.

The repeatability of the final calibrations was satisfactory. The standard deviations of the values averaged to calculate the zeros were all smaller than 0.1 Mm^{-1} which is 1% the value of light scattering by particle-free air (Rayleigh scattering). On average, the pairs of span values differed by 1% of and the largest difference was 1.7%.

The data recorded by the nephelometers were obtained by periodically dumping the digital data to a computer. The resulting files were transmitted electronically to STI, where they were assembled into the data files used for the analyses reported here. The calibration factors were applied to the measured data, and then the calibrated data were edited to delete results measured during and shortly before or after the calibrations. No other data were deleted as a result of the fact that they were outliers.

AMBIENT CONDITIONS

Figure 1 is a plot of all b_{sp} data from March 2 to 29, 2001. The b_{sp} readings exceeded 200 Mm^{-1} only for brief periods and exceeded 300 Mm^{-1} only once. Rapid changes of 100 Mm^{-1} or more were common during this time period. The decrease in b_{sp} on March 24 was very rapid. During the late afternoon, the b_{sp} readings were in the 150 to 200 Mm^{-1} range, and were approximately 190 Mm^{-1} at 1930 PST. Between 1935 and 1940 PST the b_{sp} readings decreased by about 60 Mm^{-1}, and each of the two following 5-min averages were about half the previous average. All 5-min average b_{sp} values were below 10 Mm^{-1} by 2110 PST. **Figure 2** is a plot of all RH data. Except for the last few days of the intercomparison, the RH values were high enough each night that the sample air was heated.

During fall 2001 an effort was made to obtain ambient temperature and RH data at Angiola during the time period of the intercomparison. Data for only the first 9 days of the intercomparison were available, and the RH data were valid only at a height of 2 m. The 5-min average RH data remained constant at 99.74% for 55 min during the early morning of March 3, indicating the likely presence of fog during this time period. The RH data indicate that fog was probably not present during the remainder of the 9 days with ambient RH data. However, the RH exceeded 95% during each of the remaining nights except one, when the maximum RH was 94%.

The data indicate that the intercomparison included at least one fog event that lasted approximately one hour. Also, the ambient RH was high enough every night of the intercomparison to heat the sample airflow. The daytime RH values were low enough that the sample airflow was not heated for part of each day.

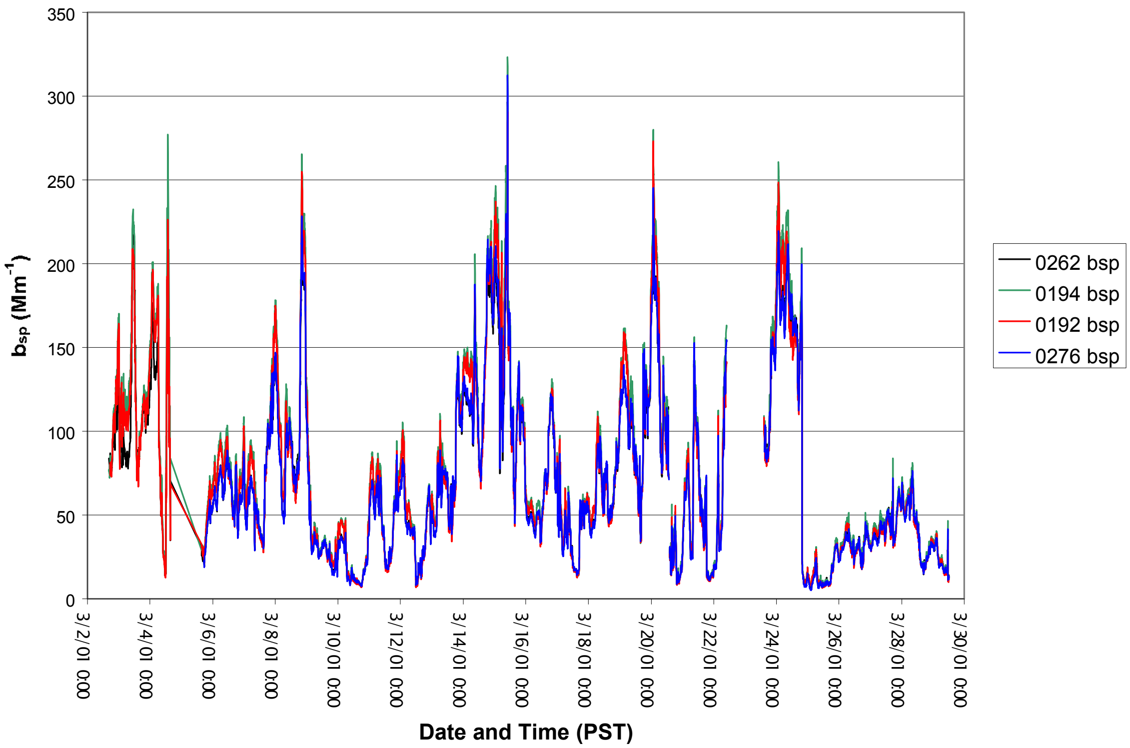

Figure 1. Time series plot of all b_{sp} data.

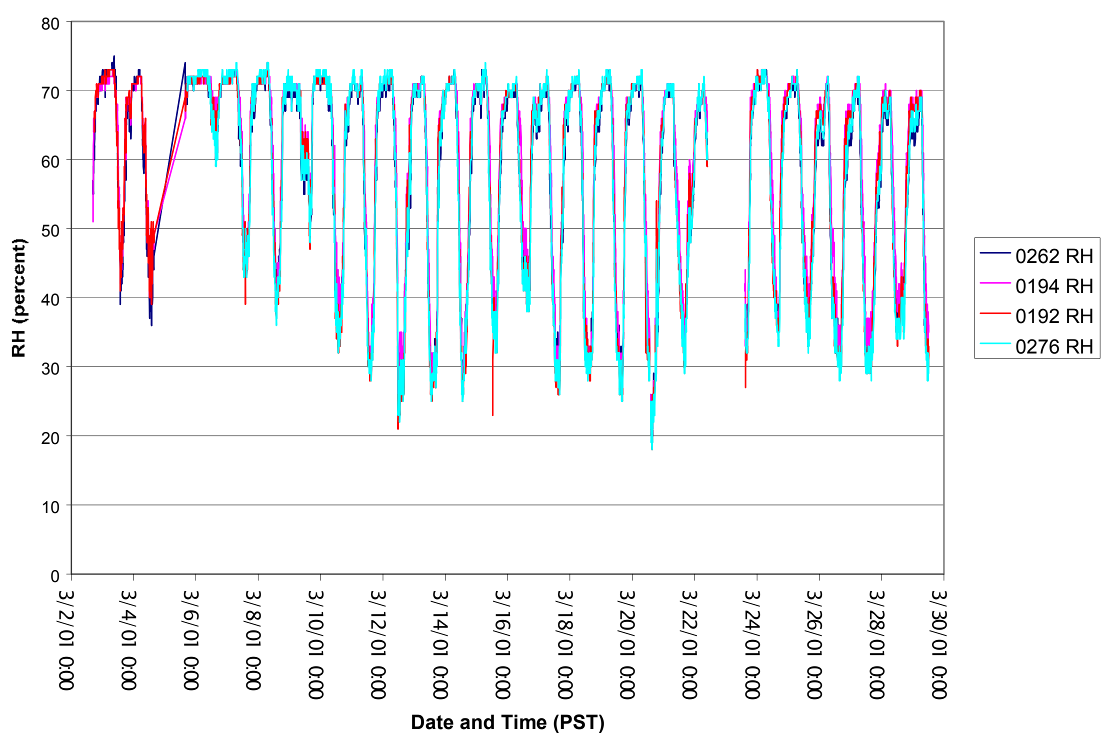

Figure 2. Time series plot of all RH data.

RESULTS

The data from pairs of nephelometers were compared using regression analysis and scatter diagrams. A preliminary analysis indicated that the relationships between the b_{sp} readings were different at low RH, when the heaters were not operating, than at high RH, when the heaters were operating. Also, intermediate relationships were observed in the transition RH range. Therefore, the data were divided into three groups. The low RH group includes all data measured when the RH recorded by nephelometer 0192 was 66% or less. The mid RH group includes all data measured when the 0192 RH was 67 to 69%, and the high RH group includes all data measured when the 0192 RH was 70% or above. The RH sensor on nephelometer 0192 was in the sample air inlet.

Figure 3 is a scatter diagram comparing b_{sp} values from nephelometers 0262 and 0276, which were both operated with the RH sensor on the sample air outlet (reversed flow) and with thermal insulation around the scattering chamber. The data in Figure 3 and the following scatter diagrams were plotted in the order indicated in the legend, and each group of data overwrites the previously plotted data. A plot of only the low RH data in Figure 3 shows a solid mass of points in the area covered by the high RH data. The agreement between this pair of nephelometers is better than for any other pair. The two outliers with b_{sp} values below 50 Mm^{-1} were recorded at 1115 PST and 1120 PST on March 29, approximately one hour before the final calibrations began. The data from all four nephelometers were disrupted during these two time periods, indicating a local source of pulses of dust. The outlier with b_{sp} values just below 100 Mm^{-1} was recorded at 1940 PST on March 24, when the b_{sp} was decreasing very rapidly. The differences in the b_{sp} readings could be caused by differences in the clocks in the nephelometers.

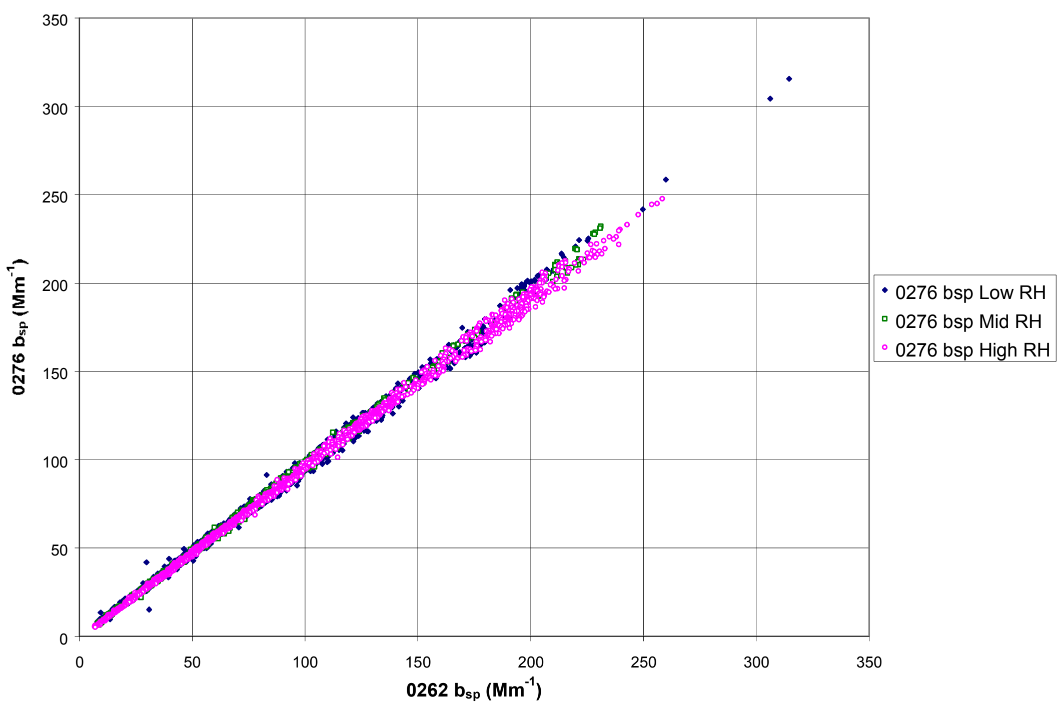

Figure 3. Scatter diagram comparing all data from nephelometers 0276 and 0262, which were operated with the RH sensor on the sample air outlet and with thermal insulation.

Figure 4 compares the b_{sp} data from nephelometers 0192 and 0194, which were operated in the original configuration with the RH sensor on the sample air inlet and no thermal insulation on the scattering chamber. The high RH data, which were plotted last and overwrite the other data, show less variability than the low RH data. The reason for the greater variability in the low RH data shown in Figure 4 compared to those shown in Figure 3 is not known. It is probable that the bias in the data in Figure 4 could be removed by more accurate calibrations of the nephelometers.

Figures 5 and 6 compare the b_{sp} data from nephelometers 0262 and 0276 respectively, which were operated with the RH sensor on the sample air outlet and with thermal insulation, with b_{sp} data from nephelometer 0192, which was operated in the original configuration. As expected, there is a bias in the data at high RH. The b_{sp} readings are higher when the RH sensor is on the sample air inlet than when the sensor is on the outlet. When heat is applied to the sample airflow, this airflow is cooled in the scattering chamber by heat lost to the air flowing through the shelter. Therefore, the air in the scattering chamber is cooler, and the RH is higher, when the RH sensor is on the nephelometer inlet than when it is on the outlet. The data in Figures 5 and 6 show that under the conditions sampled at Angiola in March 2001, this bias was small and repeatable. These data indicate that it will be possible during the analysis of the CRPAQS b_{sp} data to convert b_{sp} values measured with the RH sensor on the inlet to make them comparable to the b_{sp} values that would have been measured with the RH sensor on the outlet.

The data recorded during the fog event on March 3 were examined and it was found that they did not account for any of the outliers in Figures 5 or 6. The data recorded during the fog event were in line with other high RH data, and the outliers were recorded at other times.

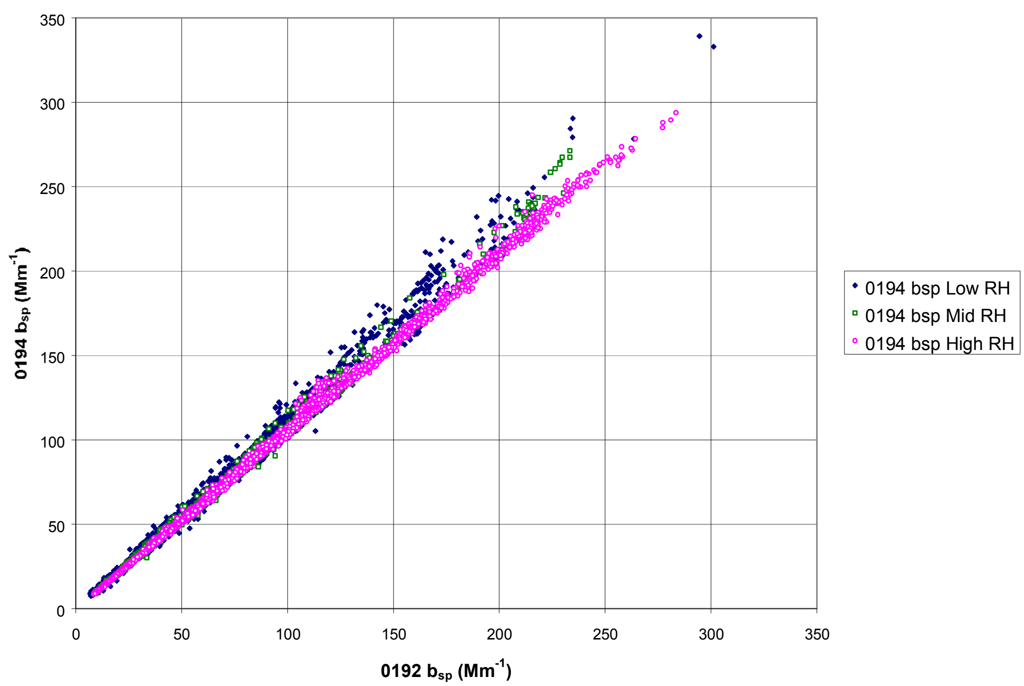

Figure 4. Scatter diagram comparing all data from nephelometers 0192 and 0194, which were operated in the original configuration with the RH sensor on the sample air inlet and no thermal insulation.

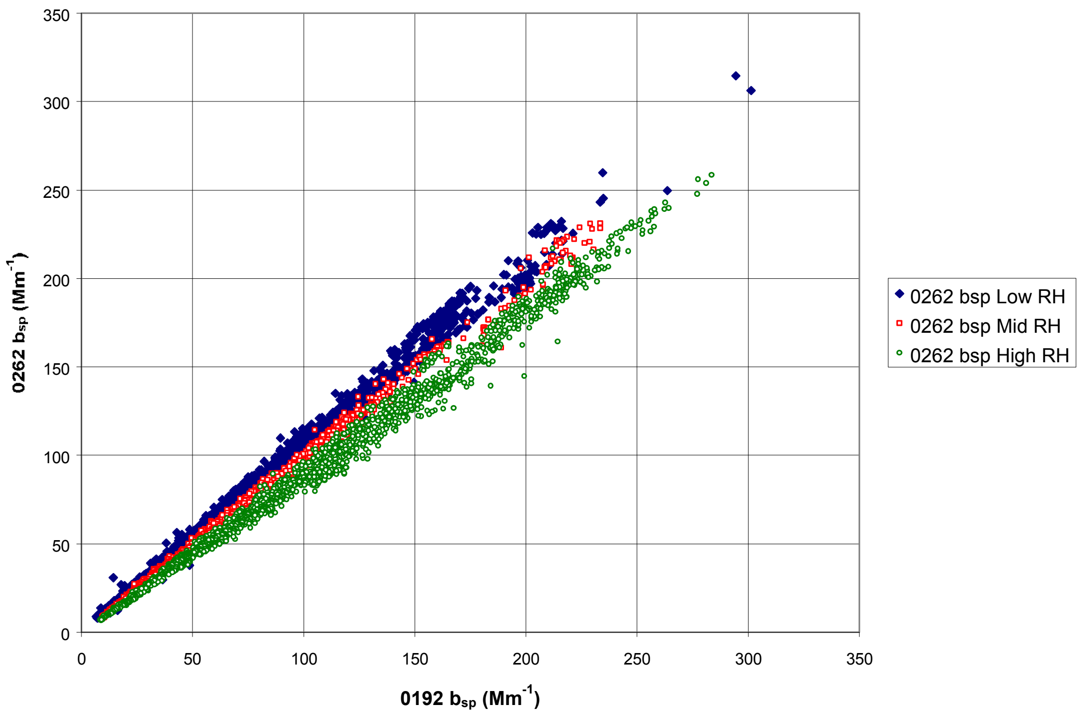

Figure 5. Scatter diagram comparing all b_{sp} data from nephelometers 0262 and 0192, which were operated with the RH sensor on the sample air outlet and inlet, respectively.

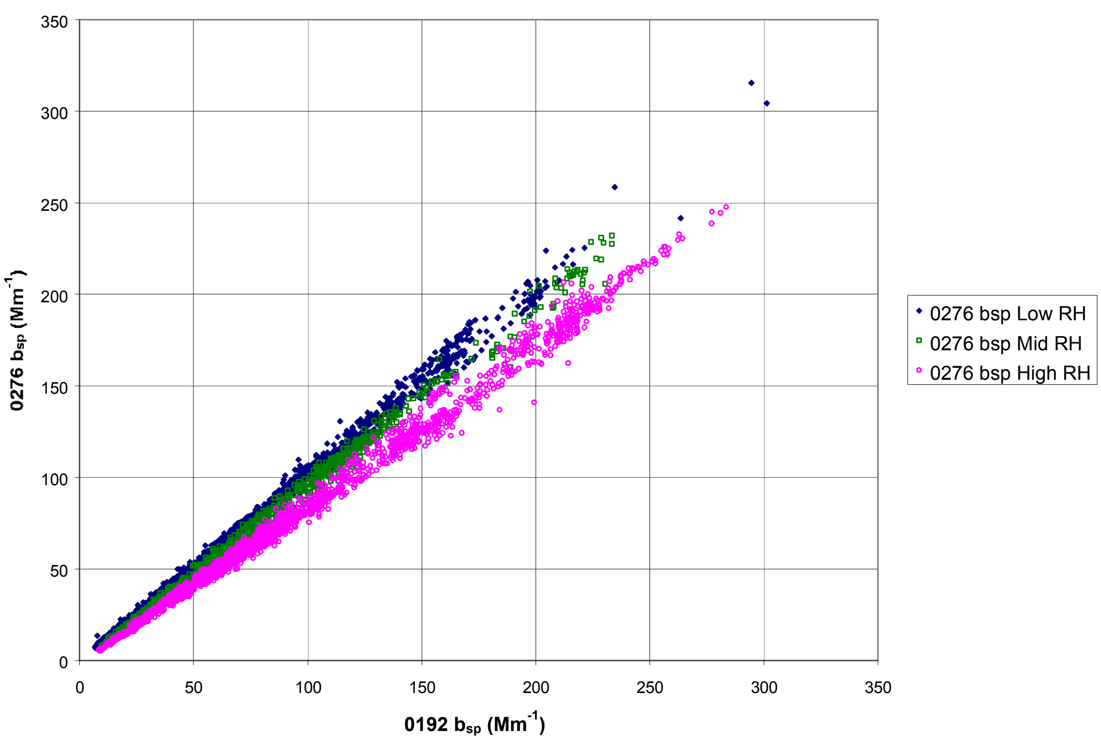

Figure 6. Scatter diagram comparing all b_{sp} data from nephelometers 0276 and 0192, which were operated with the RH sensor on the sample air outlet and inlet, respectively.

REGRESSION RESULTS

Regression analyses were performed on the data shown in Figures 3 through 6 to quantify the relations between the b_{sp} data. The results are shown in **Table 2**. As indicated above, nephelometers 0192 and 0194 were operated in the original configuration and nephelometers 0262 and 0276 were operated in the revised configuration with the RH sensor on the sample air outlet and with thermal insulation around the scattering chamber.

The data show that at low relative humidities, the b_{sp} readings from nephelometers 0192 and 0276 were quite similar, but the readings from 0276 tended to be slightly higher. The readings from nephelometer 0262 were about 3% higher than from 0276, and the readings from 0194 were nearly 10% higher than from 0192 at low RH.

The relation between the calibrations of the nephelometers can be determined from the low RH data. Under these conditions, no power is supplied to the sample air heater. The heating of the sample airflow by the nephelometer lamp and electronics is quite small, so the temperature and RH of the sample airflow in all nephelometers should be very nearly equal. At low RH values, the b_{spR} measured by nephelometers 0262 and 0276 with a reversed sample airflow is related to the b_{spI} measured by nephelometers 0192 and 0194 in the initial configuration by the linear equation.

$$b_{spR} = a + m\ b_{spI} \qquad \mathbf{(1)}$$

Equation 1 relates the calibrations of the nephelometers, and the intercept and slope are for the low RH values in Table 2. At high RH, the measurements can be related by the equation

$$b_{spR} = A + M\ b_{spI} \qquad \mathbf{(2)}$$

where the intercept and slope are for the high RH values in Table 2. The intercept and slope in Equation 2 account for both the difference in the calibration of the nephelometers and the difference in the effect of high RH values on the data. If the properly calibrated value b_{spC} for the nephelometer in the initial configuration were known, the effect of humidity could be represented by

$$b_{spR} = I + S\ b_{spC} \qquad \mathbf{(3)}$$

The low RH data provide the calibration, so b_{spC} can be calculated from Equation 1 and substituted in Equation 3 to obtain

$$b_{spR} = I + S\ (a + m\ b_{spI}) = I + a\ S + S\ m\ b_{spI} \qquad \mathbf{(4)}$$

This equation describes the high RH data, and can be compared with Equation 2 to obtain

$$M = S\ m \qquad \mathbf{(5)}$$

and

$$A = I + a\ S \qquad \mathbf{(6)}$$

Table 2. Results of the regression analysis of the b_{sp} data in Figures 3 through 6. The first two columns give the nephelometer serial number and RH range. The standard errors of the intercept and slope are reported after the plus/minus signs.

Independent (X)	Dependent (Y)	Intercept (Mm^{-1})	Slope	R Square
0262	0276 All RH	-0.82 ± 0.04	0.968 ± 0.0005	0.9992
0262	0276 Low RH	-0.64 ± 0.06	0.972 ± 0.0008	0.9990
0262	0276 Mid RH	-1.65 ± .08	0.984 ± 0.0009	0.9990
0262	0276 High RH	-1.17 ± 0.07	0.964 ± 0.0007	0.9988
0192	0194 All RH	0.24 ± 0.08	1.065 ± 0.0009	0.9955
0192	0194 Low RH	-0.31 ± 0.12	1.095 ± 0.0016	0.9928
0192	0276 Mid RH	-1.04 ± 0.17	1.078 ± 0.0019	0.9964
0192	0194 High RH	-0.28 ± 0.09	1.055 ± 0.0008	0.9980
0192	0262 Low RH	0.74 ± 0.09	1.047 ± 0.0012	0.9955
0192	0262 Mid RH	-0.35 ± 0.16	0.986 ± 0.0018	0.9962
0192	0262 High RH	-1.66 ± 0.18	0.901 ± 0.0015	0.9928
	Conversion Coeff.	-2.30	0.860	
0192	0276 Low RH	0.12 ± 0.07	1.017 ± 0.0010	0.9971
0192	0276 Mid RH	-2.04 ± 0.17	0.971 ± 0.0019	0.9958
0192	0276 High RH	-2.81 ± 0.16	0.872 ± 0.0014	0.9944
	Conversion Coeff.	-2.91	0.857	
0194	0262 Low RH	1.37 ± 0.12	0.951 ± 0.0014	0.9922
0194	0262 Mid RH	0.79 ± 0.19	0.913 ± 0.0019	0.9947
0194	0262 High RH	-1.29 ± 0.19	0.853 ± 0.0016	0.9914
	Conversion Coeff.	-2.52	0.897	
0194	0276 Low RH	0.26 ± 0.08	0.933 ± 0.0010	0.9963
0194	0276 Mid RH	-0.97 ± 0.18	0.897 ± 0.0019	0.9953
0194	0276 High RH	-2.46 ± 0.15	0.826 ± 0.0013	0.9948
	Conversion Coeff.	-2.69	0.8857	
Average	Conversion Coeff.	-2.6 ± 0.3	0.87 ± 0.02	

These equations can be solved for the coefficients in Equation 3 that describe the effect of high RH on the b_{sp} measurements

$$S = m/M \tag{7}$$

$$I = A - a\,S = A - a\,m\,/\,M \tag{8}$$

The slope S and intercept I are calculated from each of the possible pairs of nephelometers in Table 2 and the average values are tabulated at the bottom of the table. If it is assumed that all nephelometers in the field are correctly calibrated and the effect of high RH is the same as the average observed in this intercomparison, the b_{spR} that would have been observed by a nephelometer in the revised or reversed-flow configuration at RH values above 70% can be calculated from the b_{spI} measured by a nephelometer in the initial configuration (with the RH sensor on the inlet) from the equation

$$b_{spR} = -2.6\ Mm^{-1} + 0.87\ b_{spI} \tag{9}$$

This interpretation of the data was tested using the data comparing nephelometers 0276 and 0192. Equation 3 was applied to the high RH data from nephelometer 0192 using an intercept of –2.912 and slope of 0.857 calculated in Table 2. The high RH data were then compared with the low RH data as shown in **Figure 7**. The conversion brings the high and low RH data into good agreement. The agreement would have been less good if the average slope and intercept had been used for the data conversion. The bottom row in Table 2 indicates that the relative standard deviation of the values used to determine the slope in Equation 9 is approximately 2%. Therefore, the uncertainty in b_{sp} values converted using Equation 9 would be at least this large.

A conversion algorithm for RH values from 67% to 69% has not been derived and evaluated. It is expected that it would be satisfactory to use coefficients that vary linearly with RH between the low RH values and high RH values. The low RH values are $I = 0.0\ Mm^{-1}$ and $S = 1.0$ and the high RH values appear in Equation 9.

It is expected that the conversion coefficients in Equation 9 depend on the aerosol composition, which is known to vary with the season of the year. Therefore, it is possible that Equation 9 may not apply to the data from all seasons. The most likely time for variations from Equation 9 is during winter particulate matter (PM) episodes, when ambient temperatures may be low and the PM may contain higher concentrations of fine-particle nitrate and organic species than in March, when the intercomparison was performed.

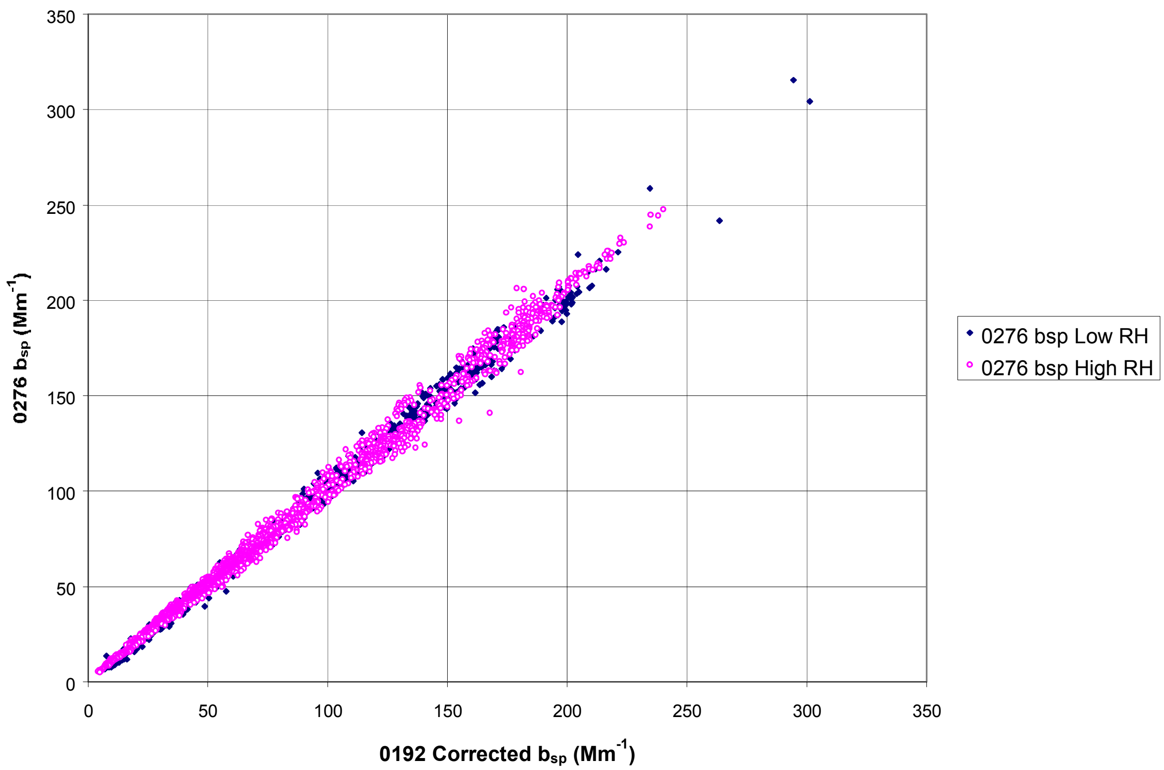

Figure 7. Comparison of high RH and low RH b_{sp} data from nephelometers 0192 and 0276 after the high RH data from nephelometer 0192 have been converted to remove the effect of RH on the measurements.

CONCLUSIONS

The scatter diagrams and regression results from this intercomparison indicate that the Radiance Research Nephelometer is capable of recording highly precise b_{sp} data. The calibration and audit data from CRPAQS indicate that the b_{sp} values measured during the daytime (when the calibrations and audits were performed) were also accurate (Richards et al., 2001).

The observation of significant differences in the b_{sp} values recorded by nephelometers in different shelters during preparations for the measurements on the tower at Angiola indicates that under some wintertime conditions, the b_{sp} values recorded by the Radiance Research Nephelometers with the RH sensor on the sample airflow inlet may not be accurate. It is recommended that b_{sp} data recorded during these preparations for the tower experiments and at other times when the RH was high and the ambient temperature was low be examined for indications of inaccurate data.

In the range of conditions encountered in this intercomparison, it is possible to convert the data measured at high RH by nephelometers in the original configuration (with the RH sensor on the scattering chamber inlet) to values that would have been measured by nephelometers in the final configuration (with added thermal insulation and the RH sensor on the scattering chamber outlet). The conversion when the measured RH is 70% or greater is given by

Equation 9. No conversion is needed at low RH, i.e., when the RH is 66% or less. It is expected coefficients in Equation 9 will vary linearly with RH for RH values from 67% to 69%.

It is expected that Equation 9 depends on the aerosol composition, and therefore may not be applicable to data from all seasons of the year. It is possible deviations from Equation 9 will occur during winter PM episodes, when ambient temperatures may be low and the PM may contain higher concentrations of fine-particle nitrate and organic species than during March, when the intercomparison was performed.

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